Kokai 06107750

IN-MOLD COATING COMPOSITION [Gatauchi hifuku soseibutsu]

Mitsutoshi Myokei et al.

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APPLICANT

(71): TITLE IN-MOLD COATING COMPOSITION (54):

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FOREIGN TITLE [54A]: Gatauchi hifuku soseibutsu <u>Claim</u> /2*

An in-mold coating composition, wherein are blended: (i) 100 parts by weight of a vehicle component comprising 20-70 parts by weight of unsaturated polyester resin, or an oligomer or resin thereof having at least 2 or more (meth)acrylate groups, and 80-30 parts by weight of copolymerizable ethylene-system unsaturated monomer; and, (ii) per [the 100 parts by weight of vehicle (i)], 1-50 parts by weight of polypropylene chloride having a chloride content of 10-50 parts by weight.

Detailed explanation of the invention

[0001]

Technical field of the invention

The present invention pertains to an in-mold coating composition that makes it possible to mold smooth films and to improve the quality of appearance, by covering defects such as pinholes and fiber patterns, for example, that are produced on the surface of fiber-reinforced plastic molded articles.

F00021

Prior art and problems to be solved by the present invention

Molded articles obtained from fiber-reinforced plastic materials in which fiber glass, carbon fibers, organic fibers, mineral fibers, for example, [are embedded in] a matrix of thermosetting resins or thermoplastic resins such as sheet molding compounds (SMC), bulk molding compounds (BMC), and stampable sheets, for example, have excellent mechanical strength and moldability, for example, and are light weight; they are widely used in fields as a material that replaces metal, such as in electrical appliance cases, automotive exterior panels, components for housing equipment, and the like.

Numbers in right margin indicate pagination of the original text.

Nevertheless, these molded articles have surface defects such as pinholes, craters, minute cracks, and fiber patterns, for example, with low luster and poor appearance, as well as [poor resistance qualities] such as poor weather resistance, water resistance, and chemical resistance, and there has been a problem with the surface hardness also being low. Accordingly, these molded articles, as normal coating, have a protective film formed on the surface, but because molded articles have surface defects as described above, even when subjected to an ordinary coating means such as spraying, for example, it is difficult for a film with excellent appearance, such as smoothness, to be formed, and the attachment characteristics are also poor; furthermore when applied to a molded article that has a thermoplastic resin for a matrix, problems exist in that solvent cracks readily form due to organic solvent in the coating material, and the coating operability is poor. Various in-mold coating methods have been proposed as methods to solve these problems. For example, such methods are disclosed in U.S. Patents 3,184,527, 4,076,788, and 4,668,460.

[0003]

Since these in-mold coating methods are specialized methods, it is difficult to use conventional coating materials [with them] as-is. Therefore recently in-mold coating compositions have been developed that are suitable for in-mold coating methods; the following in-mold coating compositions may be mentioned as examples: those disclosed in Japanese Patent No. Sho 54[1979]-13273, Sho 59[1984]-15137, and Sho 59[1984]-19583. These in-mold coating compositions cover the surface defects of molded articles and have good coating operability, but the smoothness of the obtained film is unsatisfactory, and there is a problem in particular with the attachment to fiber reinforced plastic molded articles that have a matrix of thermoplastic resin. Keenly aware of this state of affairs, the inventors performed diligent research, which resulted in the discovery of a in-mold coating composition that used

the superior points of conventional in-mold coating compositions to good advantage, and that also formed a film with excellent [properties] such as smoothness and adhesion, for example.

[0004]

Means to solve the problems

More specifically, the present invention offers an in-mold coating composition, wherein are blended:

(i) 100 parts by weight of a vehicle component comprising 20-70 parts by weight of unsaturated polyester resin, or an oligomer or resin thereof having at least 2 or more (meth)acrylate groups, and 80-30 parts by weight of copolymerizable ethylene-system unsaturated monomer; and, (ii) per [the 100 parts by weight of vehicle (i)], 1-50 parts by weight of polypropylene chloride having a chloride content of 10-50 parts by weight. The following statements explain the present invention [in greater detail].

[0005]

The in-mold coating composition that is used in the present invention is constituted from: (i) a vehicle component comprising unsaturated polyester resin, or an oligomer or resin thereof having at least 2 or more (meth)acrylate groups, and ethylene-system unsaturated monomer; (ii) polypropylene chloride; (iii) ordinarily used pigment; (iv) polymerization initiator; and (v) mold parting agent; and also including various types of additives and modified resins, for example, that may be added as the occasion requires. Specific examples of the aforementioned oligomer or resin thereof having at least 2 or more (meth)acrylate groups that constitutes [part of] (i), the vehicle component, include oligomers or liquid resins that are ordinarily used as vehicle components of in-mold coating compositions, such as epoxy (meth)acrylate, urethane (meth)acrylate, polyester (meth)acrylate, and polyether (meth)acrylate, as well as mixtures of 2 or more of these

[0006]

In general it is preferred that the weight average molecular weight of these oligomers and their resins, while differing depending on the type, is approximately 300-10,000. The oligomer or resin thereof has at least 2 or more, preferably from 2-4, (meth)acrylate groups per 1 molecule of oligomer or resin thereof. The following statements provide a more detailed explanation of the oligomer or resin thereof of the present invention. The aforementioned epoxy acrylate compound is a compound that has been manufactured by a common ring-opening addition reaction of acid onto the epoxy group, using an epoxy compound and unsaturated carboxylic acid in proportions such that the carboxyl group equivalent weight is 0.5-1.5 per 1 equivalent weight of epoxy group. Acrylic acid and methacrylic acid may be mentioned as representative examples of the aforementioned unsaturated carboxylic acid. Bisphenol A-type epoxy and phenolic novolac-type epoxy, for example, may be mentioned as representative examples of epoxy compounds.

[0007]

The aforementioned urethane (meth)acrylate compound is manufactured by a method known to the art with the proportion of organic diisocyanate and organic diol having a hydroxyl group, as well as hydroxyalkyl (meth)acrylate, being such that the NCO/OH ratio is 0.9-1.0; for example, it may be manufactured by producing a polyurethane prepolymer having terminal isocyanate [groups] by organic diol and organic diisocyanate in the presence of a urethane catalyst such as dibutyltin dilaurate, for example, and then effect a reaction of hydroxyalkyl (meth)acrylate until almost all the free isocyanate groups are reacted. The proportion of organic diol and hydroxyalkyl (meth)acrylate is suitably 0.1-0.5 moles of the former per 1 mole of the latter. As the aforementioned organic diisocyanate it is possible to

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use those organic diisocyanates that are ordinarily used in paints, such as toluene diisocyanate, isophorone diisocyanate, and polymethylene polyphenyl diisocyanate, for example; the 2,4- and 2.6-isomers of toluene diisocyanate are particularly useful.

[0008]

The following may be mentioned as typical examples of the aforementioned organic diol: alkylene glycols such as ethylene glycol, propylene glycol, diethylene glycol, polyethylene glycol, and polypropylene glycol, for example, and diester diols that are the diester reaction products of dicarboxylic acids and their anhydrides. The compound expressed by the general formula

$$CH_2 = CRCO_2 - (C_nH_{2n})-OH$$

(where R is -H or CH₃, and n is an integer from 2-8) is useful as the aforementioned hydroxyalkyl (meth)acrylate. The aforementioned polyester acrylate is the reaction product that is manufactured by a reaction between the aforementioned unsaturated carboxylic acid and a polyester polyol having a hydroxyl group on the terminal. The aforementioned polyester polyol is typically obtained by an esterification reaction of a saturated or unsaturated dicarboxylic acid or acid anhydride thereof with an excess amount of alkylene diol. The following may be mentioned as typical examples of the aforementioned dicarboxylic acid: oxalic acid, succinic acid, adipic acid, phthalic acid [and] maleic acid; and as typical examples of the aforementioned alkylene diol there may be mentioned ethylene glycol, propylene glycol, butane diol, and pentane diol.

[0009]

The aforementioned polyether acrylate is a compound that is manufactured by a reaction between a polyether polyol such as polyethylene glycol or polypropylene glycol and the aforementioned unsaturated carboxylic acid. The suitable weight average molecular weight of these oligomers and resins thereof is approximately 300-10,000, preferably 500-5,000. As the aforementioned unsaturated polyester resin that constitutes [part of] vehicle component (i), it is possible to use that which is ordinarily used as vehicle components for in-mold coating composition; it is manufactured by effecting a publicly known reaction between an organic polyol and an unsaturated polycarboxylic acid, and if required by effecting the reaction of a saturated polycarboxylic acid. The following may be mentioned as typical examples of the aforementioned organic polyol: ethylene glycol, propylene glycol, triethylene glycol, trimethylol propane, glycerine, bisphenol A, and bisphenol S. The following may be mentioned as typical examples of the aforementioned unsaturated polycarboxylic acid: (anhydrous) maleic acid, fumaric acid, (anhydrous) itaconic acid. The suitable average molecular weight of the unsaturated polycarboxylic acid is approximately 800-10,000, preferably 1,000-4,000.

[0010]

It is also possible to use both the unsaturated polyester resin and the oligomer or resin thereof having at least 2 or more (meth)acrylate groups. Typical examples of the aforementioned ethylene-system unsaturated monomer that constitutes [part of] vehicle component (i) include: styrene, alpha-methylstyrene, cross-styrene, vinyl toluene, divinyl benzene, methyl (meth)acrylate, ethyl (meth)acrylate, propyl (meth)acrylate, ethylene glycol (meth)acrylate, and trimethylolpropane tri(meth)acrylate, but it is not limited to these.

[0011]

The vehicle component (i) comprises the aforementioned unsaturated polyester resin, or an oligomer or resin thereof having at least 2 or more (meth)acrylate groups, and the aforementioned copolymerizable ethylene-system unsaturated monomer; the ratio for blending these together will depend on the specific types thereof, but ordinarily 20-70:80-30 (weight basis) is suitable, because within this range it is possible to obtain a in-mold coating composition having suitable hardness properties and viscosity. The polypropylene chloride (ii) that is used in the present invention is blended in order to improve the attachment and smoothness of the film formed on the surface of the molded article. The polypropylene chloride used in the present invention is a polymer in which a propylene homopolymer or a copolymer of butene or ethylene, for example, containing approximately 70 mol % or more of propylene is chlorinated, and the [suitable] number average molecular weight is approximately 5000-50,000, preferably 10,000-40,000. The number average molecular weight is set in this way because there is a tendency for smoothness to be degraded if it is less than 5000, and a tendency for compatibility with the aforementioned vehicle component (i) to decline if it exceeds 50,000. The chlorine content of the polypropylene chloride (ii) is 10-50 wt%, preferably 20-45 wt%. If the chlorine content exceeds the aforementioned range, the attachment of the film will be degraded, and if it is less the storage stability of the in-mold coating composition will be poor and it will be difficult to obtain a smooth film, which are both disadvantageous. The amount of polypropylene chloride to be blended is 1-50 parts by weight, preferably 2-30 parts by weight, per 100 parts by weight of the aforementioned vehicle component (i); within this range it is possible to obtain a film with excellent smoothness and attachment properties, for example. It is undesirable for [the amount] to exceed this [range], because attachment and weather resistance, for example, will be degraded.

[0012]

As the pigment (iii) to be used in the present invention, it is possible to use various colored pigments and extender pigments that have been conventionally ordinarily used for plastics and for paints. Typical examples of specific colored pigments include: titanium oxide for white; benzidine yellow, titanium yellow, and Hansa yellow for yellow; molybdate orange, chrome yellow, benzi[di]ne orange for orange; quinacridon and maroon for red; chrome green, chrome oxide green for green; ultramarine, cobalt blue, and ultramarine [sic] for blue; carbon black, black iron oxides or powdered pigments such as iron oxide, for example, and scalar pigments such as mica, for example, that have been treated with nickel, aluminum, graphite, titanium oxide, and the like, for black. The following may be mentioned as representative examples of extender pigments: calcium carbonate, talc, barium sulfate, aluminum hydroxide, silica, and clay, for example. Note that the pigment is blended for the objects of coloring the molded article and effecting an attractive appearance, causing dispersal of the contraction stress that accompanies film hardening, improving attachment with the base material (molded article), filling the craters that are still present on the molded article surface, smoothing minute irregularities (waviness) on the surface, and improving the appearance of the molded article.

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[0013]

The amount of pigment to be blended for this purpose corresponds to 10-150 parts by weight per 100 parts by weight of the aforementioned (i) component. It is not necessarily necessary to blend colorant/pigment if a top coat of colorant paint is applied to the obtained film or if a clear finish [is desired]. The polymerization initiator (iv) that is used in the present invention produces free radicals, and is used for the purpose of effecting polymerization of the aforementioned vehicle component. The following may be mentioned as typical examples of specific polymerization initiators: tertiary butyl peroxybenzoate, tertiary butyl peroxy-2-ethyl hexanoate, tertiary butyl peroxyisopropyl carbonate, lauroyl peroxide, tertiary butyl peroxylaurate, 1,1-bis(tertiary butyl peroxy)-3,3,5-trimethyl cyclohexane, and acetyl acetone peroxide.

[0014]

A suitable blending proportion of the polymerization initiator is 0.2-10 parts by weight per 100 parts by weight of the aforementioned (i) vehicle ingredient. The mold parting agent (v) used in the present invention is added in order to effect the smooth separation of the hardened film from the mold, and the following may be mentioned as typical examples of its types: stearates of zinc, aluminum, magnesium, and calcium, for example, and alkyl phosphates, for example. The suitable blending proportion [of the mold parting agent] is 0.1-10 parts by weight per 100 parts by weight of the aforementioned (i) vehicle ingredient. The invented in-mold coating composition has the above-explained ingredients (i)-(v) as structural ingredients, and as the occasion may require, it is further structured by the blending of various additives such as curing promoters, dispersants, anti-settling agents, fluidizing promoters, polymerization inhibitors, and ultraviolet adsorbers, for example, modified resins such as polymethylmethacrylate resin and saturated polyester resin, and plasticizers, for example.

[0015]

The following statements explain a method for in-mold coating that utilizes the invented in-mold coating composition. The fiber reinforced plastic molding material used in the present invention are conventionally publicly known [resins] such as SMC, BMC, FRTP, and stampable sheets, for example, and these can be utilized without any particular limitations. The following may be mentioned as typical specific examples: [namely] the aforementioned fiber reinforced plastic molding materials that have as the matrix a thermosetting resin such as an unsaturated polyster resin-system, epoxy acrylate resin-system, phenolic resin system, and epoxy resin systems; or a thermoplastic resin such as a polyolefin resin-system, polystyrene resin-system, polyocarbonate resin-system, polybutylene

terephthalate resin system, polyester resin, or polyphenylene oxide resin-system, for example. Conventional methods for molding inside a metal mold may be used without any particular limitations as the molding method, but the method described in Japanese Patent No. Sho 61[1986]-273921 and Patent No. Sho 55[1980]-9291 are particularly suitable. More specifically, the aforementioned fiber reinforced plastic molding material is placed into a mold that is formed with a cavity space, in which the shape of the intended molded article has been formed by placing one mold within another (hereinunder, for the sake of convenience they will be called the "upper mold" and the latter the "lower mold"); they are joined, and [the molded article] is formed within the mold. More specifically, when the molding material has a thermoplastic resin as the matrix, the inside of the mold is subjected to heating and/or pressurization, the molding material is caused to flow [into the mold] while at the same time a hardening reaction is allowed to occur, thus forming the intended shape. The molding heating temperature is to be determined optionally depending on the molding time and the type of molding material, for example, but ordinarily 130-200°C is suitable; it is preferred that the mold be already set to the aforementioned temperature prior to the introduction of the molding material, and that this temperature be maintained until the hardened film described below is obtained.

[0016]

The molding pressure is to be determined optionally depending on the heating temperature and the type of molding material, for example, but ordinarily 50-200 kgf/cm² is suitable. The molding time period may [extend] until the thermosetting reaction of the molding material is complete, and may be described as [the length of time required beginning from] when the in-mold coating composition is introduced, [until] it becomes the degree of hardness such that there is no loss to the shape of the molded article, and ordinarily 40-200 sec is suitable. On the other hand, when the molding material has a

thermoplastic resin as the matrix, the material that has been softened by being pre-heated, for example in a heating oven, is subjected to pressurization in the mold so as to form the molding material into the intended shape, and the in-mold coating composition is introduced, [so in this case] it is caused to harden to a strength that does not impair the shape of the molding entity. After the molding entity has been hardened in this fashion, the upper mold is separated from the surface of the molding entity, and [something] is larger than the desired hardening film thickness to be described below; but after a gap has been provided that is insufficient to permit the separation of the aforementioned mold joining, or after maintaining the aforementioned molding pressure without conferring a gap and in a condition where the molds are joined, or after the aforementioned pressure is depressurized, the in-mold coating composition is introduced (injection introduction) into the space between the upper mold and the surface of the molding entity in an amount just sufficient to give a hardened film of the intended film thickness, preferably 30-1000 microns.

[0017]

Then, when the molding material has a matrix of thermosetting resin, the heating temperature is maintained at a predetermined temperature, the coating composition is coated evenly onto the surface of the molded article, this is subjected to a (repeat) pressurization of approximately 20-150 kgf/cm² so as to cause penetration, and this is normally maintained for approximately 20-240 sec, until a hardened film has been formed. On the other hand, when the molding material has a matrix of thermoplastic resin, the molded article is not re-softened, [but] the mold temperature is maintained at the temperature for hardening the coating composition, for example 60-160°C, the coating composition is coated evenly onto the surface of the molded article, this is subjected to a (repeat) pressurization of approximately

20-150 kgf/cm² so as to cause penetration, and this is normally maintained for approximately 20-240 sec, until a hardened film has been formed. In this case the combined use of an accelerator, such as cobalt naphthenate or amines, for example, is preferred, because it is desirable for the hardening temperature of the coating composition to be low. After a hardened film has been formed on the molded article surface in this way, the mold is opened, and the molded article is taken out, thus giving a molded

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[0018]

Effect of the Invention

article with a protective coating.

The invented in-mold coating composition makes it possible to form a film with excellent [properties] such as smoothness and attachment, and that covers the surface defects such as pinholes of the surfaces of fiber reinforced plastic molded articles.

Application examples

The following statements describe the present invention in more detail by reference to application examples. In the application examples, "parts" and "%" are by weight.

Application Examples 1-3 and Comparative Examples 1-5

In-mold coating compositions were prepared by kneading together and dispersing the ingredients shown in Table 1 (excluding the polymerization initiator); immediately prior to their use the polymerization initiator was added.

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	カーボンブラック	7	8	8	7	7	7	7	8
	タルク	60	60	40	60	60	60	69	40
	ステアリン酸亜鉛	0.5	0.7	0.7	0.5	0.5	0.5	0.5	0.7
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						(1	Unit:	parts	s)

Key: 1 Application Examples

- 2 Comparative Examples
- 3 Urethane acrylate

Epoxy acrylate

Unsaturated polyester resin

Styrene

Hydroxypropyl methacrylate

Trimethylol propane trimethacrylate

Polypropylene chloride

Polypropylene

Polypropylene chloride

Polypropylene chloride

Carbon black

Talc

Zinc stearate

8% cobalt octylate

Modified resin

Tertiary butyl peroxybenzoate

4 Note

[0020]

Note 1: polyethylene glycol - tolylene diisocyanate - hydroxyethylene acrylate oligomer: weight average molecular weight 2700; 2 acrylate groups per 1 molecule

Note 2: epoxy compound ["Epicoat 828" (Yuka Shell Epoxy)] - methacrylate oligomer: weight average molecular weight 540; 2 methacrylate groups per 1 molecule

Note 3: the reaction product of a polybasic acid comprising 1 mole maleic anhydride and 1 mole isophthalic acid with a polyhydric alcohol comprising 1 mole neopentyl glycol and 1 mole propylene glycol; number average molecular weight 1500

Note 4: chlorine content of 41%, number average molecular weight of 35,000

Note 5: chlorine content of 0%, number average molecular weight of 30,000

Note 6: chlorine content of 60%, number average molecular weight of 40,000

Note 7: chlorine content of 25%, number average molecular weight of 40,000

Note 8: vinyl acetate resin

[0021]

Molded articles were prepared that had films according to the molding apparatus and molding method of Japanese Patent No. Sho 61[1986]-273921, using the in-mold coating composition obtained in Application Examples 1-3 and Comparative Examples 1-5. This method is explained in detail as follows. A chrome-plated flat experimental mold 300 mm long and 300 mm wide was used, and this was set to a mold temperature of 120°C. First, a thermoplastic glass fiber reinforced plastic material sheet (fiberglass content of 40%), which had a matrix of polypropylene resin, was heated to approximately 200°C in a heating oven and said sheet was placed on a lower mold; this was then molded under the conditions of molding pressure of 150 kgf/cm² and molding time of 30 sec. The molding pressure was then reduced to 20 kgf/cm², 19 ml of the aforementioned in-mold coating composition was introduced into the space between the upper mold and the molding entity, a pressure of 70 kgf/cm² was again applied, and this was held for 180 sec. Then the mold was opened, and the coated molded article, coated with a film of approximately 200 microns film thickness, was taken out. The obtained coated molded article was subjected to tests for film appearance, surface roughness, attachment, and hot water resistance, for example, and the results of these tests are shown in Table 2. In Table 2 it is clearly shown that the films obtained in Application Examples 1-3, in which the invented in-mold coating composition was used, have excellent appearance, smoothness, attachment, and hot water resistance. In contrast, in Comparative Example 1, in which non-chlorinated polypropylene was used, and in Comparative Example 5, which did not contain polypropylene chloride, have no commercial value, because the film was partially taken into the mold, so an even film was not obtained. In Comparative Example 2, in

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which polypropylene chloride was used that had a large chlorine content, and in Comparative Example 3, in which the amount of [pollypropylene chloride was small, had low scores for film appearance, attachment, and hot water resistance. The water resistance was poor in Comparative Example 4, which had an excessive content of polypropylene chloride.

[0022]

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Key: 1 Application Examples

2 Film appearance

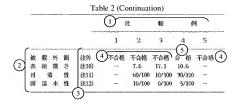
Surface roughness

Attachment

Hot water resistance

- 3 Note
- 4 Pass

[0023]



Key: 1 Comparative Examples

2 Film appearance

Surface roughness

Attachment

Hot water resistance

- 3 Note
- 4 Fail
- 5 Pass

[0024]

Note 9 [Film appearance]: 2-liquid type urethane coating was coated by spraying onto the coated molded article to a dry film thickness of 35 ± 5 microns, this was dried at 80° C x 30 min, and the appearance was evaluated by visual observation. Pass: no abnormalities on film. Fail: abnormalities such as cracks or fractures occur on the film.

Note 10 [Surface roughness]: according to JIS B 0601-1982. Unit: Rmax [microns]

Note 11 [Attachment]: coated molded articles coated with a top-coat, as obtained in Note 9, were subjected to a peel test with a cellophane tape cut into 2 mm wide lattice pattern, and the number of remaining squares were counted. The number of remaining squares was not counted for objects that had peeled from the molded article undercoat and objects when only the top coat film was peeled.

Note 12 [Hot water resistance]: An attachment test identical to that of Note 11 was performed on coated molded articles coated with a top-coat, as obtained in Note 9, that had been immersed in 40°C hot water for 240 h and then allowed to stand at room temperature for 1 h.

[0025]

Application Examples 4-6 and Comparative Examples 6-9

In-mold coating compositions were prepared by kneading together and dispersing the ingredients shown in Table 3 (but excluding the polymerization initiator); immediately prior to their use the polymerization initiator was added.

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エボキシアクリレート	注2	32	30		32	32	32	33	
ポリエステル	注15)		30						
アクリレート									
スチレン (4)	30	30	40	30	30	30	30	
とドロキシプロピル		6	10	10	6	6	6	(
メタクリレート									
塩素化ポリプロピレン	(E4)	2	2	40			60		
ボリアロゼレン	注5)				2				
塩素化ポリプロピレン	注6)					2			
数化チタン		23	20	30	20	20	20	20	
タルク		20	20	20	20	26	20	25	
炭酸カルシウム		60	60	60	60	60	69	66	
ステアリン酸亜鉛		0.6	3	0.6	0.6	0.6	0.5	0.6	
8%オクテル酸コバルト		3	1	1	3	1	1	1	
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Key: 1 Application Examples

- 2 Comparative Examples
- 3 Urethane acrylate

Epoxy acrylate

Polyester acrylate

Styrene

Hydroxypropyl methacrylate

Polypropylene chloride

Polypropylene

Polypropylene chloride

Titanium oxide

Talc

Calcium carbonate

Zinc stearate

8% cobalt octylate

Acetyl acetone

Tertiary butyl peroxybenzoate

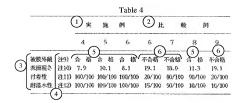
4 Note

[0026]

Note 15: "Photomer 5018" (San Nopco Limited); weight average molecular weight 600; 4 acrylate groups per 1 molecule

As in Application Example 1, a thermoplastic glass fiber reinforced plastic material sheet (fiberglass content of 35%), which had a matrix of polybutylene terephthalate resin, was heated to approximately 300°C, and said sheet was placed on the lower mold of a mold that had been set to 130°C; this was then molded under the conditions of molding pressure of 200 kgf/cm² and molding time of 30 sec. The molding pressure was then cancelled, 10 mL of the aforementioned in-mold coating composition was introduced into the space between the upper mold and the molding entity, a pressure of 60 kgf/cm² was again applied, and this was held for 90 sec. Then the mold was opened, and the coated molded article, coated with a film at approximately 100 microns thickness, was taken out. The obtained coated molded article was subjected to the same tests as in Application Example 1, and the results of these tests are shown in Table 4.

[0027]



- Key: 1 Application Examples
 - 2 Comparative Examples
 - 3 Membrane appearance

Surface roughness

Adhesion properties

Hot water resistance

- 4 Note
- 5 Pass
- 6 Fail